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REPRESENTATIONS OF 3-D SCATTERING MATRICES

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The standard Mueller matrix formalism in which the incident light beam and all scattered light beams lie in a single plane is extended to 3-D geometry. This study shows that there are two matrix representations, which are appropriate for describing light scattering in unrestricted directions, that are related to each other by the rotation matrix. One matrix is better suited for characterization of the scatterer, and the other performs the traditional Mueller matrix function of relating outgoing Stokes vectors to the incident Stokes vector.

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PREFACE

The work described in this report was authorized under Project No. 1L162622A552, Smoke and Obscurants. This work was started in June 1989 and completed in July 1989.

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CONTENTS

LIST OF FIGURES

REPRESENTATIONS OF 3-D SCATTERING MATRICES

1. INTRODUCTION

Aerosols play an increasingly significant role in the way the military views its battlefields. Airborne dusts impair the ability of the eye and the electronic sensor to perceive danger or acquire targets. Dusts may be generated incidentally as by-products of military activity (e.g., battlefield road dusts, smokes from burnings), or they may be intentionally delivered as screening smokes. The early detection of deadly chemical or biological agents that might be dispersed in the air is clearly a matter of enormous importance to the U.S. Army.

In any study of aerosols and their effects, it is necessary to characterize the aerosol in question [e.g., to describe the range of sizes and shapes of the constituent particles, their concentration, the material(s) of which they are made, and the changes these properties undergo as the cloud develops and ages]. Inelastic (i.e., no change in wavelength) light scattering is an important technique among those available for aerosol characterization. Broadly speaking, an aerosol is illuminated by a light beam, and inferences about the aerosol are drawn according to the nature and spatial distribution of the resulting scattered light. Numerous applications of this technique have been found because of its versatility and speed. Light scattering can be used to scrutinize a single aerosol particle or probe clouds many kilometers thick; with it, one can study samples on hand in the laboratory or particulates remotely distant in the atmosphere or even in interstellar dust clouds.

The U.S. Army Chemical Research, Development and Engineering Center (CRDEC) Nephelometry Laboratory was set up to explore fresh ways of applying light scattering techniques to aerosol characterization. For example, the advent of screening smokes effective in infrared and millimeter-wave regions of the spectrum has been based on using highly nonspherical particles (flakes and fibers). This resulted in a need to replace traditional instruments for sizing spherical particles with new methods capable of analyzing irregular particles. Over the years, CRDEC's attention has been primarily focused on finding the size and shapes of aerosol particles. To that end, we have been particularly interested in the description and measurement of the polarization states of light and in trying to relate polarization transformations to characteristics of the aerosol. In modern optical parlance, the polarization state of a light beam is summarized by a set of four numbers known as the beam's Stokes vector. A process that alters the Stokes vector is described with the Mueller matrix, which is a 4 by 4 matrix relating incident and altered (or scattered) Stokes vectors.

In this report, we review the standard Stokes/Mueller formalism applicable to scattering in a plane and then extend those ideas to scattering in three dimensions (i.e., the case when a description is required for light scattered simultaneously into all directions about a particle).

2. STOKES VECTORS

All experimentally accessible information needed to characterize the intensity and state of polarization of a nearly monochromatic light beam is contained in the Stokes vector, the four elements of which are called Stokes parameters. Several different sets of optical quantities can serve as Stokes parameters for describing light, and at least three have been suggested over the years. In this report, the term "Stokes vector" refers to the system most commonly used today, usually denoted {I, Q, U, V,} and discussed by Van de Hulst,[1] and Bohren and Huffman,[2] and by Kerker[3] who prefers the symbols $\{S_0, S_1, S_2, S_3\}$.

Mueller's phenomenological definition of the Stokes parameters was reported by Parke,[4] one of his students. For the experimentalist, Mueller's definition has the compelling attraction of referring only to observable quantities. In fact, Mueller's definition of Stokes parameters is a prescription for measuring the parameters; conversely, the numerical values one obtains upon carrying out the prescribed measurements on a beam of light are the Stokes parameters of that particular beam. Mueller's definition may be described by referring to Figure 1.

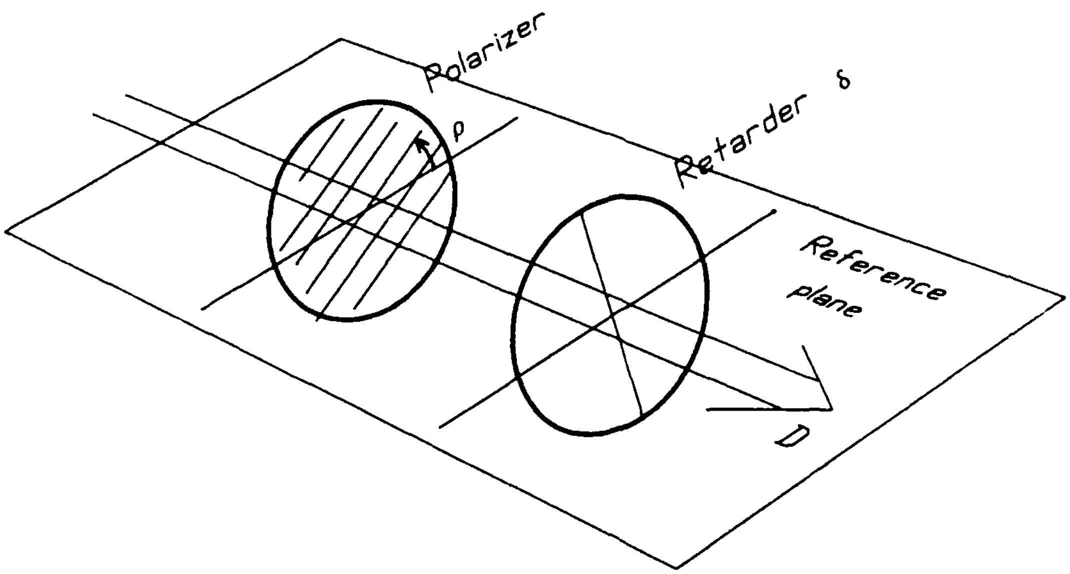


Figure 1. Optical Elements Required to Determine the Stokes Parameters of a Light Beam

In Figure 1, a reference plane containing the parallel beam of light in question is specified, and the beam intensity is measured with detector D. The beam passes through an ideal polarizer whose transmission axis is at an angle ρ with respect to the reference plane. The beam may also transit an ideal wave plate of retardation δ, whose preferred axes are parallel and perpendicular to the reference plane. (Mueller's phenomenological approach is carefully self-consistent but is not reproduced here. Terms such as "ideal polarizer," "ideal wave plate," "transmission axis," etc., and even "parallel beam of light" are defined operationally.)

The detected intensity in the setup of Figure 1 can be described by the following formula:

$$\mathrm{Int} = 1/2\ [I + Q \cos 2\rho + (U \cos \delta + V \sin \delta) \sin 2\rho] \tag{1}$$

A sequence of measurements with various polarizer angles (ρ = 0°, 90°, 45°, -45°) and phase shifts [δ = 90°, 0° (i.e., wave plate removed)] will determine the Stokes parameters of the beam, I, Q, U, and V (Mueller referred to them as I, M, C, and S).

Today, Stokes parameters corresponding to the set defined by Mueller are usually expressed in terms of the electric field components of a plane wave satisfying Maxwell's equations. If the electric field of a simple plane wave is written as follows:

$$E = \mathrm{Re}\ [E_\ell \mathbf{l} + E_r \mathbf{r}] \tag{2}$$

where E_ℓ and E_r are the complex, oscillating field components parallel to and perpendicular to the reference plane (r x l in the direction of propagation z), Mueller's Stokes parameters can be shown to be the following real quantities:

$$\begin{aligned} I &= E_\ell E_\ell^* + E_r E_r^* \\ Q &= E_\ell E_\ell^* - E_r E_r^* \\ U &= E_\ell E_r^* + E_r E_\ell^* \\ V &= i(E_\ell E_r^* - E_r E_\ell^*) \end{aligned} \tag{3}$$

Many find it easier to visualize the field in terms of real amplitudes and phases. If one writes the following equations,

$$\begin{aligned} E_\ell &= a_\ell\, e^{-i(kz + \epsilon_\ell)}\, e^{i\omega t} \\ E_r &= a_r\, e^{-i(kz + \epsilon_r)}\, e^{i\omega t} \end{aligned} \tag{4}$$

then the Stokes parameters are expressed as follows:

$$
\begin{aligned}
I &= a_\ell^2 + a_r^2 \\
Q &= a_\ell^2 - a_r^2 \\
U &= 2a_\ell a_r \cos\delta \\
V &= 2a_\ell a_r \sin\delta
\end{aligned}
\qquad (5)
$$

where $\delta = \epsilon_\ell - \epsilon_r$, the phase difference between parallel and perpendicular components whose amplitudes are a_ℓ and a_r.

The Stokes vectors of equations 3 and 5 are not identical to the operationally defined Stokes vector. Equations 2 and 4 describe only a 100% polarized plane wave satisfying $I^2 = Q^2 + U^2 + V^2$; actual light is often quite different. Real light is modeled as a superposition of simple plane waves (not necessarily all identical) arriving in rapid succession. The theoretical Stokes vectors are then modified by including a time average of sufficient length to smooth the instantaneous field fluctuations as follows:

$$
\begin{aligned}
I &= \langle E_\ell E_\ell^* + E_r E_r^* \rangle = \langle a_\ell^2 + a_r^2 \rangle \\
Q &= \langle E_\ell E_\ell^* - E_r E_r^* \rangle = \langle a_\ell^2 + a_r^2 \rangle \\
U &= \langle E_\ell E_r^* + E_r E_\ell^* \rangle = \langle 2a_\ell a_r \cos\delta \rangle \\
V &= i\langle E_\ell E_r^* - E_r E_\ell^* \rangle = \langle 2a_\ell a_r \sin\delta \rangle
\end{aligned}
\qquad (6)
$$

Then, as in the Mueller definition, $I^2 \geq Q^2 + U^2 + V^2$ and $Q = U = V = 0$ correspond to completely nonpolarized (natural) light. Notice that the definitions of equation 6 involve theoretical quantities that cannot be directly observed (field amplitudes and phases) and that fluctuate much too rapidly to track and average, even inferentially from intensity measurements. However, electromagnetic theory is indisputable, and definitions in terms of its elements are indispensable for making theoretical predictions of the Stokes parameters and the matrices that connect incoming and outgoing Stokes vectors across an optical instrument or scattering region.

Whatever is taken as the primary definition of the Stokes vector, a reference plane must always be specified, for either resolving fields into orthogonal parallel and perpendicular components or serving as a reference direction for setting optical elements. Even when the Stokes vector is unambiguously defined, the values of its parameters Q and U are not intrinsic properties of the radiation alone but depend on this reference plane. The selection of a reference plane is quite arbitrary with respect to the light source, except that it must contain the beam in question. Usually, it is chosen in a manner that is natural for the experimental setup contemplated or used and often is simply the table top onto which the equipment is mounted.

If the Stokes parameters of a beam are known with respect to some reference plane, then the Stokes vector referenced to any other plane containing the beam can be calculated by applying the rotation matrix **R** (ϕ).

A light beam directed into the reader's eye is indicated in Figure 2. The beam's Stokes vector should be $S_0 = \{I, Q, U, V\}$ with respect to the reference plane P. A new reference plane P results from rotating P' counterclockwise about the beam by a (positive) angle ϕ The Stokes vector referenced to the new plane, $S' = \{I', Q', U', V'\}$, is given by equation 7:

$$S' = R(\phi)\ S_0 \tag{7}$$

or, explicitly by equation 8:

$$\begin{bmatrix} I' \\ Q' \\ U' \\ V' \end{bmatrix} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos 2\phi & \sin 2\phi & 0 \\ 0 & -\sin 2\phi & \cos 2\phi & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix} \tag{8}$$

Notice that I (intensity), V (dominance of right circular polarization over left circular polarization), and the quantities $Q^2 + U^2 + V^2$ (a measure of the degree of polarization) are invariant under reference frame rotations.

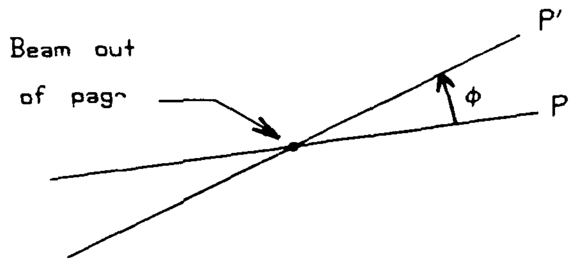


Figure 2. Rotation of the Reference Plane

3. MUELLER MATRIX

Nearly all interactions of light with matter, and particularly all the elastic scattering processes with which this report is concerned, are linear in nature. When a beam of light with Stokes vector S_0 is incident on an optical instrument or scattering system, any emerging beam S can be related

to the incident beam by a 4 by 4 matrix **M** whose elements are independent of the intensity.

$$\mathbf{S} = \mathbf{M}\,\mathbf{S}_0 \tag{9}$$

M is called the Mueller matrix; its purpose is to allow the prediction of **S**, given any $\mathbf{S}_0$.

Although the concept of Mueller matrices can be very useful for following the polarization changes experienced by an essentially unscattered beam of light transmitted or reflected by the elements of an optical instrument, we are only concerned with light scattering phenomena and will henceforth restrict the discussion to such cases in this report.

The two Stokes vectors $\mathbf{S}_0$ and **S** are always defined with reference to the plane that contains them both. The numerical values of the 16 matrix elements in any instance depend on the configuration of the scattering system, its orientation in the reference plane, and the directions of $\mathbf{S}_0$ and **S**. The use of the Mueller matrix is illustrated in Figure 3; a light beam is incident on a scattering object, and we consider light scattered into a narrow cone at 25° off the incident direction.

The Mueller matrix **M** in Figure 3 was made up for illustrative purposes and tells us, for instance, that if horizontally polarized light of unit intensity strikes the scatterer ($\mathbf{S}_0 = \{1, 1, 0, 0\}$), then the outgoing light at 25° will have the Stokes vector $\mathbf{S} = \{0.013, 0.010, 0.001, 0.000\}$, which is considerably diminished in intensity and no longer purely horizontally polarized, although nearly so. The same set of numbers in **M** operates on any incident Stokes vector $\mathbf{S}_0$.

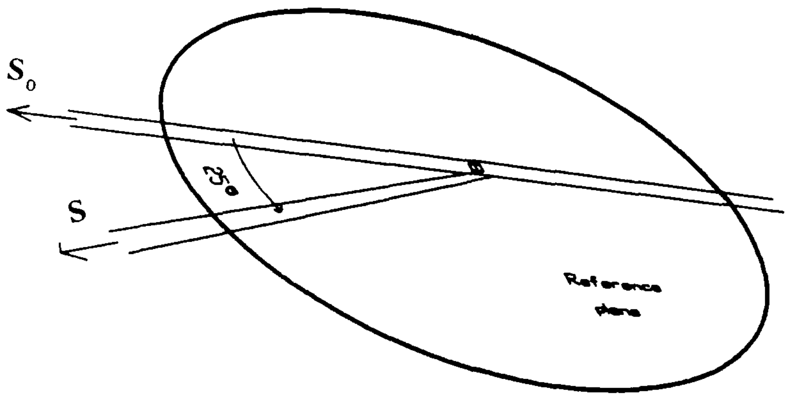
It is often convenient to express scattering in terms of the normalized Mueller matrix (indicated in Figure 3), which is derived by dividing each element of the Mueller matrix by the 1, 1 element. The 1, 1 element is taken outside the matrix as a multiplying scalar; the remaining elements of the normalized matrix all lie in the [-1, 1] range.

Mueller sought to formulate all concepts of optics on the basis of empirical laws and devised an operational definition for the matrix that transforms Stokes vectors. In Mueller's definition, a scatterer is illuminated in turn by four light beams prepared in four independent polarization states, and the Stokes parameters of the corresponding scattered beams are measured. The Mueller matrix elements are then constructed from combinations of incident and scattered beam Stokes parameters.

The Mueller matrix may also be defined in terms of wave theory concepts by considering the most general transformation of a simple wave. If an incident wave is represented with the complex electric field components $E_{\ell 0}$ and E_{r0}, as in equation 4, and if only linear processes are involved, then the emerging or scattered wave is most generally represented by E_ℓ and E_r, where

$$
\begin{aligned}
E_\ell &= A_2\, E_{\ell 0} + A_3\, E_{r0} \\
E_r &= A_4\, E_{\ell 0} + A_1\, E_{r0}
\end{aligned}
\tag{10}
$$

Van de Hulst[1] shows how to write each of the 16 elements of the Mueller matrix in terms of the four complex coefficients A_i above, and points out that because there are only eight constants represented by the A's (including one irrelevant phase), there can be only seven independent constants contained in the Mueller matrix. The nine relations that must therefore exist among the 16 Mueller matrix elements were not worked out by Van de Hulst; however, they have been derived and published by Fry and Kattawar.[5]



$$
M(25°) = \begin{pmatrix} 0.010 & 0.003 & 0.001 & 0.000 \\ 0.003 & 0.007 & -0.002 & 0.000 \\ -0.001 & 0.002 & -0.004 & 0.003 \\ 0.000 & 0.000 & -0.003 & -0.004 \end{pmatrix} = 0.01 \begin{pmatrix} 1.0 & 0.3 & 0.1 & 0.0 \\ 0.3 & 0.7 & -0.2 & 0.0 \\ -0.1 & 0.2 & -0.4 & 0.3 \\ 0.0 & 0.0 & -0.3 & -0.4 \end{pmatrix}
$$

Figure 3. Mueller Matrix Predicts the Scattered Stokes Vector Produced by Any Incident Stokes Vector

Of course, not all scattering systems consist of rigid immutable objects as was supposed above. For example, aerosol particles are in constant motion, rotating, tumbling, and shifting positions relative to each other. The Stokes vector of light scattered from a dynamic particle (or an assembly

of particles) fluctuates in concert with the particle motions and may be regarded as "produced" by a Mueller matrix with correspondingly rapidly fluctuating elements. Most experiments that measure Mueller matrix elements of fast dynamic systems will report independently time-averaged values for each of the 16 elements because the scattering system ranges rapidly many times over its allowed values during the course of a single measurement.

Fry and Kattawar[5] also treated this time-averaged case (viewed equivalently as an incoherent sum of Mueller matrices) in their paper and showed that each of the nine equations relating quadratic functions of Mueller matrix elements, which they had derived for the simple stationary case, either failed completely or at best became one-way inequalities when averaging had to be taken into account. Therefore, in this most general case, all 16 Mueller matrix elements are independent. (However, the number of independent elements may still be reduced to fewer than 16 even in a dynamic or multiparticle scattering system, depending on symmetries exhibited by the individual particles. Van de Hulst[1] or Perrin[6] provide further details.) The distinction drawn between the Mueller matrices of stationary and dynamic systems is analogous to the earlier distinction between Stokes vectors of simple plane waves (wherein $I2 \geq Q2 + U2 + V2$) and Stokes vectors of superpositions of plane waves ($I2 \geq Q2 + U2 + V2$).

The light scattering instruments developed for CRDEC – the Boeing Multichannel Nephelometer (Boeing Company, Seattle, WA) and the Wyatt Submicron Particle Analyzer (Wyatt Technology Corporation, Santa Barbara, CA) – do not make time averaged intensity measurements. The instruments sample light scattered by individual particles in a laser beam through time windows so short (microseconds) that there can be no perceptible change in the particle's orientation during data acquisition.

Scattering problems are frequently concerned with how an optically observable quantity, such as intensity, is distributed as a function of scattering angle. Mueller matrix elements are observable quantities; in fact, taken together, they comprise a complete description of the scattering properties of a system. Figure 4 shows the angular dependence of the normalized Mueller matrix elements corresponding to a homogeneous sphere with a size parameter $X = \left(\frac{2\pi r}{\lambda}\right) = 3.600$ and a complex refractive index ($N = n - i\kappa$) given by $n = 1.500$ and $\kappa = 0.01$.

The 16 matrix element values were calculated at each degree of scattering angle in the range of 0°, 180°. The plots in Figure 4 are in positions that correspond to the matrix elements displayed on them (i.e., the plot located in the first row and second column shows $M_{1,2}$ versus θ. The 181 discrete values in each plot have been joined point-to-point by straight lines to give the appearance of continuous curves.

Although such graphs are loosely called Mueller matrices, Figure 4 is actually a representation of many different but related Mueller matrices (181 matrices in this example), all corresponding to the same sphere but connecting incident to outgoing Stokes vectors at 181 different directions.

The Mueller matrix formalism was invented as a convenient mean of predicting and summarizing the action of a scatterer or an instrument on a beam of light. However, the Mueller matrix, especially an elaborate representation of multiple matrices such as shown in Figure 4, can play a second role by serving as a descriptor of the scattering system. In that role, the Mueller matrix may constitute a valuable method for particle characterization.

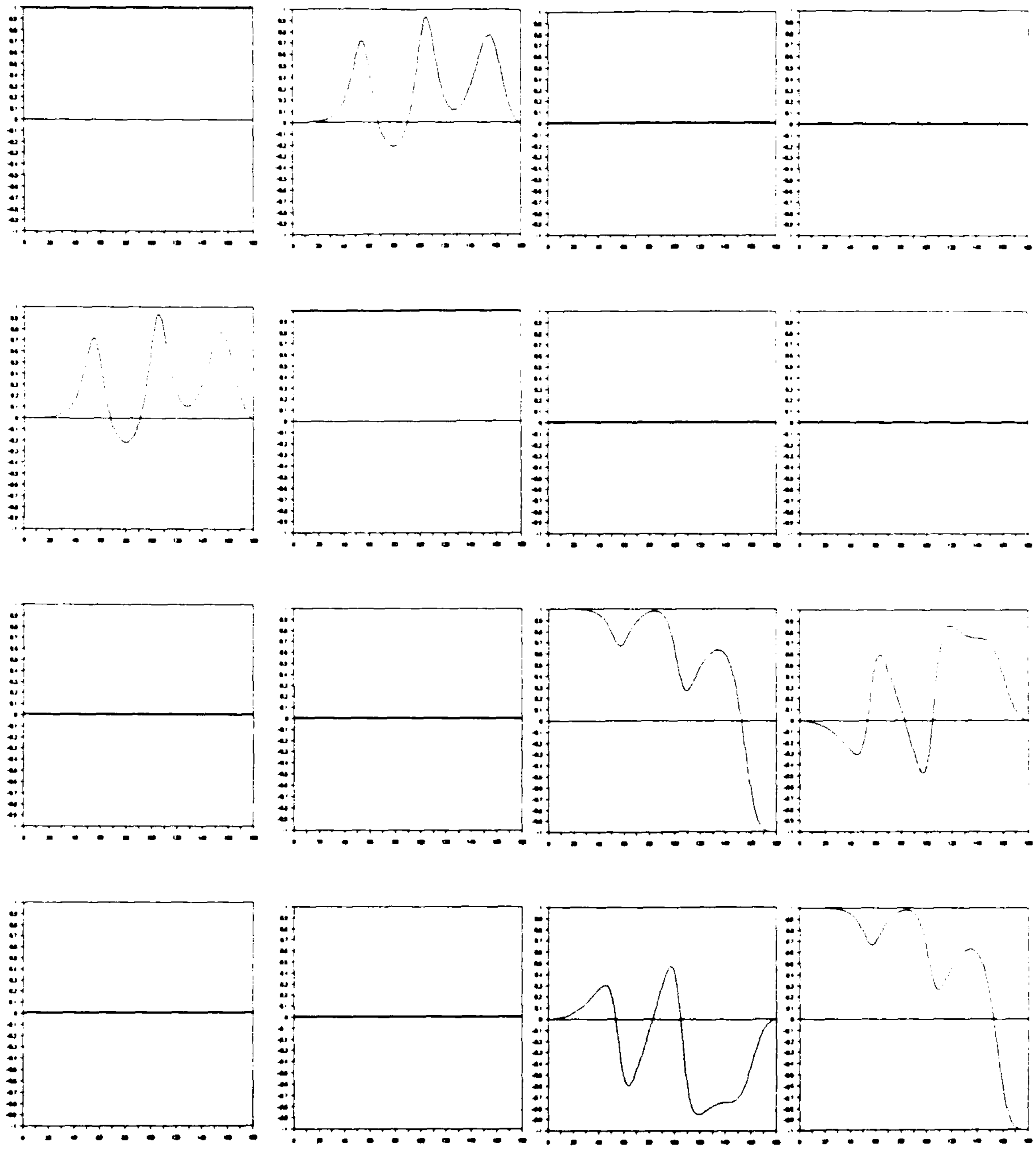


Figure 4. Sixteen Normalized Mueller Matrix Elements as Functions of Scattering Angle for a Sphere of Size Parameter 3.60, $n = 1.50$, $\kappa = 0.01$

To the slightly practiced eye, Figure 4 reveals information about its associated scatterer. The facts that at all scattering angles $M_{2,2} = 1$, $M_{1,2} = M_{2,1}$, $M_{3,4} = -M_{4,3}$, $M_{3,3} = M_{4,4}$ and the upper light and lower left quadrants vanish are all consistent with a spherical scatterer. The sphere's size parameter can be roughly estimated by counting the number of relative minima (or maxima) across any of the oscillating matrix elements. In fact, it is probable, but not certain (uniqueness questions are still quite unresolved in this area) that only a sphere with $X = 3.6$, $n = 1.5$, and $\kappa = 0.01$ is consistent in every detail with the Mueller matrices represented in Figure 4. Therefore, in principle, measuring the Mueller scattering information of Figure 4 is tantamount to identifying the scatterer as a sphere with the stated properties.

4. 3-D MUELLER MATRICES

We can attempt to identify a sphere by its Mueller matrices because we expect a one-to-one correspondence between the sphere's physical properties and its set of matrix values. There can be only one set of Mueller matrices associated with any given sphere, and presumably no other scatterer has exactly the same matrix set as that sphere. However, this correspondence is lost in the scattering of nonspherical particles. The pattern of light scattering around an irregular particle depends on the particle's orientation in the scattering plane. Therefore, there will be as many different sets of Mueller matrices (i.e., Figure 4) associated with any one irregular particle as there will be ways to twist and turn its orientation.

To sidestep this problem and make at least some progress beyond spheres, we first consider light scattering only from particles with fixed orientations. Then, we may be able to at least postulate a one-to-one relationship between the shape of an oriented irregular particle and its Mueller matrices; but, the situation is still more complicated than with spherical scatterers. Unlike the case with spheres, Mueller matrices belonging to an irregular particle generally differ from one plane to another because the particle's appearance varies with the plane in which it is viewed, and the scattering in one plane is not predictable from the scattering in another. Even for a fixed particle, knowledge of the light scattering in a single plane is only a small portion of the information available for its characterization; therefore, we must enlarge the Mueller matrix concept to include all directions about a scatterer.

The extension of the planar representation of Mueller matrices with elements $M_{i,j}(\theta)$ to a 3-D representation $[D_{i,j}(\phi, \theta)]$ is straightforward, assuming that the planar Mueller matrices are available by calculation or measurement in any desired plane through the scatterer.

Looking at the calculation first, suppose a particle whose Mueller matrix is calculable in any orientation is fixed in some reference frame, as in Figure 5. First, we calculated the Mueller matrix elements for a number of scattering angles (in the 0-180° range) in that reference plane, whose azimuth (ϕ) was taken to be 0°. In other words, we tabulate values for $D_{i,j}(0°, \theta)$.

The incident Stokes vector and all scattered Stokes vectors connected by Mueller matrices calculated in this fashion are referenced to the $\phi = 0$ plane. So far, this is the sort of data displayed in Figure 4.

Next, we changed our vantage point to a different half plane corresponding to a different azimuth angle (i.e., for instance $\phi = 10°$ in Figure 5). In this plane, the particle has a different orientation; it is rotated by -10° about the incident beam compared to its appearance in the $\phi = 0°$ plane. We again calculated the Mueller matrix elements (for the "new" scatterer) for scattering in the $\phi = 10°$ plane and obtained the connection between incident and scattered Stokes vectors that all referred to the $\theta = 10°$ azimuth plane $[D_{i,j}(10°, \theta)]$.

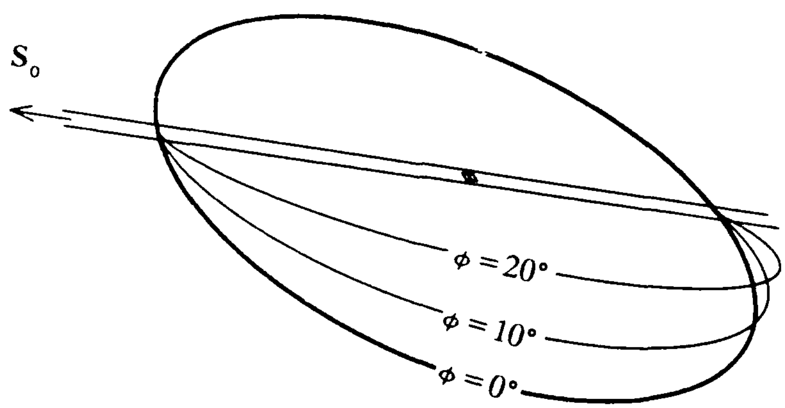


Figure 5. Calculating Matrix Elements Along Many Scattering Planes Passing Through the Incident Beam

We repeated this procedure in many half planes about the incident beam (in the example in Figure 5) at 20°, 30°, etc., around to 350°. In each case, we calculated the Mueller matrix elements as functions of scattering angle for the particle viewed in that particular azimuth plane.

To display this calculated data, we constructed a 3-D graph for each of the Mueller matrix elements, plotting the element along the z-axis above an x-y plane scaled by scattering angle [0°, 180°] and azimuth angle [0°, 360°]. To illustrate such plots, we considered scattering from the homogeneous sphere of Figure 4 and calculated and plotted its Mueller matrix elements as described above. The results are shown in Figure 6.

A single slice through all 16 plots at $\phi = 0°$ is the exact equivalent of the data in Figure 4. There is no ϕ dependence in this special case because a sphere looks the same no matter how it is rotated. The pattern in the plane $\phi = 0°$ is simply repeated at all other azimuth angles. All special characteristics (noted in Figure 4) that revealed the scatterer to be a sphere ($D_{2,2} = 1$ everywhere and so on) are still plainly evident.

[The data in Figures 6 and 7 were plotted using OMNIPLOT, a commercial graphics software package for IBM compatible PCs. Values of z (matrix element) are plotted for θ from 0° to 180° in increments of 5°, for ϕ from 0° to 360° in steps of 10°, and those values are connected by lines making a fishnet display. The plot is then converted to a solid display by removing hidden lines. A perimeter of four lines with $z = 0$ has been added around the actual data points to aid visualization of positive and negative matrix values as a sort of trampoline display. Other artifacts are spikes of +1 and -1 in opposite corners added to force OMNIPLOT's automatic scaling to produce plots of uniform appearance and to aid in visualization of the data.]

While a sphere may seem a poor example with which to illustrate creating a 3-D representation because there is no ϕ dependence, it is a very good choice to illustrate the following point: The **D** matrix data displayed in Figure 6 are wrong, at least in the traditional sense of what a Mueller matrix does. We require a matrix that will predict the Stokes vector of every scattered light beam, given the Stokes vector of any incident beam. The matrix **D** fails this test.

According to the data in Figure 6, the matrix elements of **D** are independent of azimuth angle ϕ. If it were true that at every angle $\mathbf{S}_{out} = \mathbf{D}\mathbf{S}_{in}$, then the outgoing Stokes vectors $\mathbf{S}_{out}$ would also be independent of ϕ; this is generally not true, even for a spherical scatterer. For example, consider a specific incident beam that is linearly polarized parallel to the reference plane $\phi = 0°$ (i.e., the beam is horizontally polarized). Its Stokes vector is, to within a constant, given by $\mathbf{S}_{in} = \{1, 1, 0, 0\}$. The light scattered at any scattering angle θ_0 in the $\phi = 0°$ plane is also horizontally polarized ($\mathbf{S}_{out} \propto \{1, 1, 0, 0\}$). However, light scattered from the same incident beam through an angle (including θ_0) in the $\phi = 90°$ plane must be polarized perpendicularly to that 90° plane. Its Stokes vector will be proportional to $\{1, -1, 0, 0\}$ and cannot be the same as the Stokes vector at the same scattering angle in the $\phi = 0°$ plane, as indicated in Figure 6.

The problem is that in calculating the 3-D representation from a sequence of 2-D representations, we have introduced a multiplicity of reference planes without taking their effect into account. In the calculation, we assumed that at each azimuth plane both the scattered and incident Stokes vectors were referenced to that plane. It is true that each scattered beam's

Stokes vector must be referenced to the particular azimuth plane in which the beam lies, but the incident beam lies in all the azimuth planes and can be referenced to any of them. At each step in the calculation leading to Figure 6, we allowed the incident Stokes vector to be redefined, depending on which plane we were in. However, at the end, we talk of the incident Stokes vector, meaning only the Stokes vector referenced to the plane $\phi = 0°$.

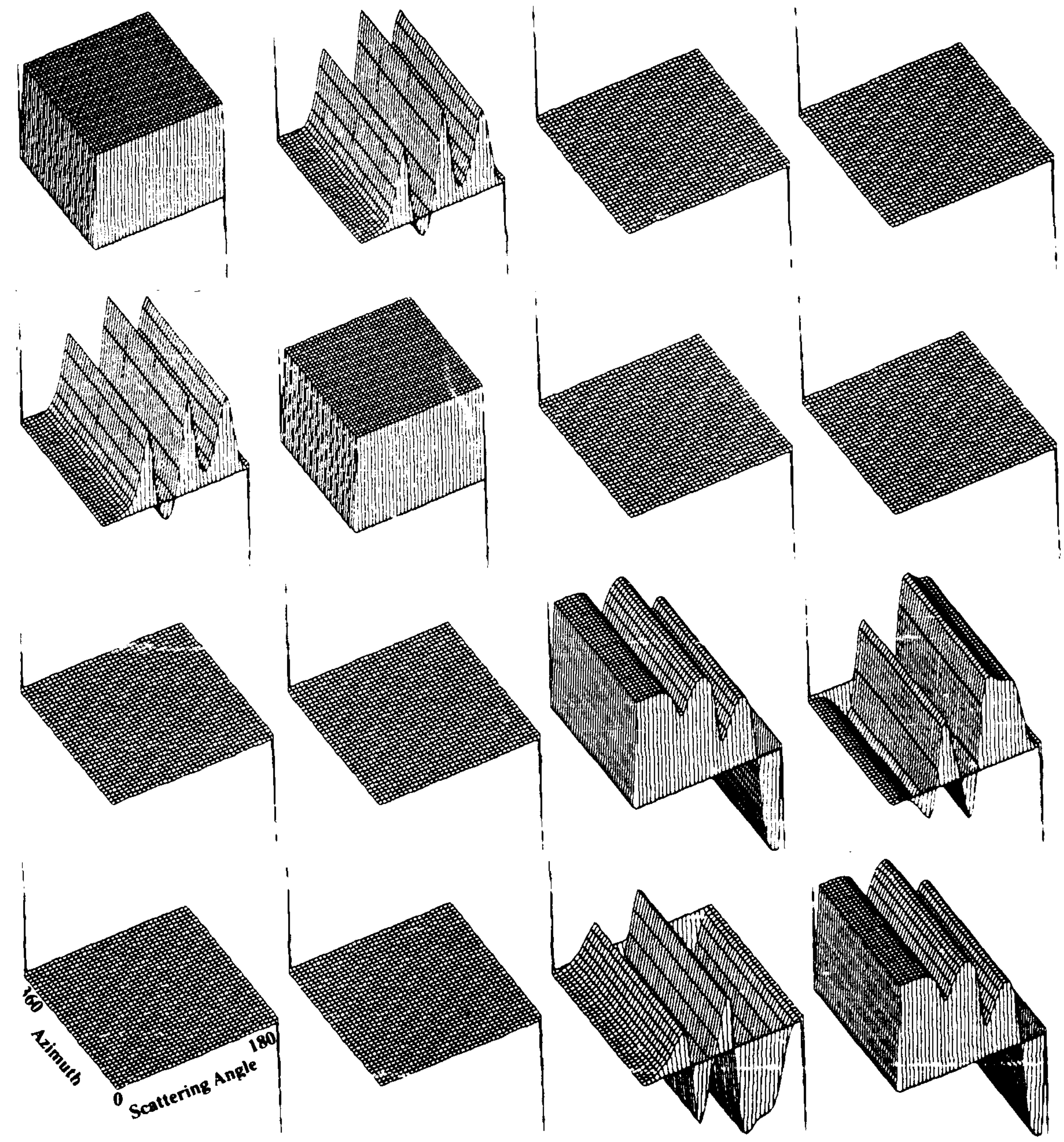


Figure 6. Representation of the D-Matrix Elements as Functions of the Scattering Directions ϕ and θ (Azimuth and Scattering Angle) for the Sphere of Figure 4

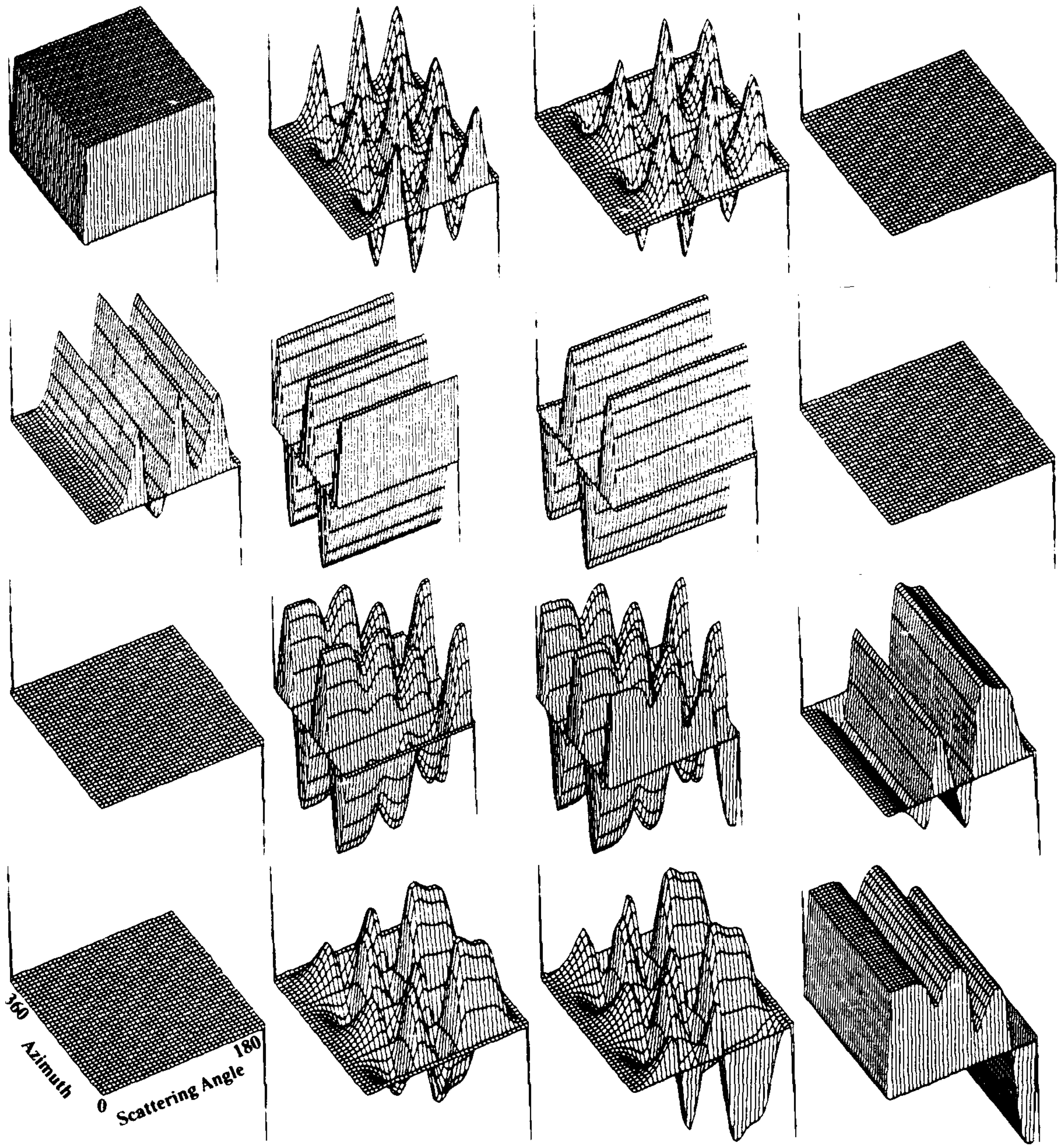


Figure 7. Representation of the Mueller Matrix Elements as Functions of the Scattering Directions ϕ and θ (Azimuth and Scattering Angle) for the Sphere of Figure 4

The solution is to repeat the calculation and explicitly rotate the incident Stokes vector (originally referenced to the $\phi = 0°$ plane) to its representation in any other ϕ plane before performing the Mueller matrix calculation in that plane. For example, beams scattered into the $\phi = 10°$ plane from an incident beam S_0 (referenced to the $\phi = 0°$ plane) are given by equation 11:

$$S(10°,\theta) = \mathbf{D}(10°,\theta)\ \mathbf{R}(10°)\ \mathbf{S}_0 \tag{11}$$

because $\mathbf{R}$ (10°) $\mathbf{S}_0$ is the incident Stokes vector expressed in the $\phi = 10°$ plane.

It is the product of **D** and **R** that serves as the Mueller matrix description of the scattering event in the sense of predicting all the scattered beams for any one given incident beam. We call that product $\mathbf{M}$ (ϕ, θ).

$$\mathbf{M}(\phi,\theta) = \mathbf{D}(\phi,\theta)\ \mathbf{R}(\phi) =$$

$$\begin{array}{llll} D_{1,1} & (D_{1,2}\cos2\phi - D_{1,3}\sin2\phi) & (D_{1,2}\sin2\phi = d_{1,3}\cos2\phi) & D_{1,4} \\ D_{2,1} & (D_{2,2}\cos2\phi - D_{2,3}\sin2\phi) & (D_{2,2}\sin2\phi = d_{2,3}\cos2\phi) & D_{2,4} \\ D_{3,1} & (D_{3,2}\cos2\phi - D_{3,3}\sin2\phi) & (D_{3,2}\sin2\phi = d_{3,3}\cos2\phi) & D_{3,4} \\ D_{1,4} & (D_{4,2}\cos2\phi - D_{4,3}\sin2\phi) & (D_{4,2}\sin2\phi = d_{4,3}\cos2\phi) & D_{4,4} \end{array} \tag{12}$$

Performing the multiplications indicated above, we obtained the data in Figure 7 as the "correct" Mueller matrix elements for the $x = 3.60$ size parameter sphere. However, those relationships and symmetries that helped identify the scatterer as a sphere in the **D** matrix display (Figure 6) do not carry over into the Mueller matrix display, **M**, in Figure 7. It is no longer true at all scattering directions that $M_{2,1} = 1$, $M_{3,4} = -M_{4,3}$, $M_{2,1} = M_{1,2,}$, $M_{3,3} = M_{4,4}$; and half of the upper right and lower left quadrant elements no longer vanish.

The price for obtaining the functionally correct Mueller matrix is an increased complexity in its graphical representation increased in fact to a degree even greater than that suggested by comparing Figures 6 and 7. Equation 12 shows that elements from the second and third columns of **D** are mixed (i.e., summed) in the matrix **M**. It so happens that for spheres, one member of each of the pairs $D_{i,2}$ and $D_{i,3}$ is always zero; therefore, the transformation from **D** to **M** only involves multiplying the second and third column elements of **D** by $\cos2\phi$ or $\sin2\phi$. However, for nonspherical scatterers, that simplification will not hold and a most intractable pattern can be expected to emerge in the second and third columns of **M**, even for relatively simple shapes.

It was pointed out in Section 3 that the 2-D representation of Mueller matrices served two purposes: (1) the data predicted the outcome of light scattering experiments, and (2) the data characterized the scattering

particles. In the 3-D case, we see that those two functions are best performed by two different matrices. The **D** matrices should be plotted when a description or characterization of the scattering particle is required; whereas, the Mueller **M** matrices predict how incident light will scatter from the particle. The two matrices are related by equation 12.

Computer codes are available for calculating the 2-D scattering matrix (as a function of θ) for a few basic nonspherical particles such as spheroids, cylinders, cubes, etc. If a 3-D matrix is required for such a particle, then **D** would be the natural matrix to calculate because to obtain it the 2-D case would simply be applied over and over while incrementally rotating the particle about the incident beam axis. The elements of **M**, if needed, could later be computed from the elements of **D**. On the other hand, laboratory experiments such as those at CRDEC, designed to detect and measure particle scattering properties on several different planes simultaneously, will yield data directly in terms of the Mueller matrix **M** (ϕ, θ). If particle characterization is the ultimate goal of the measurements, then elements of **D** should be computed from the measured data.

Finally, we note that eight elements of the first and last columns are the same in both **M** and **D** representations. This is a cogent reason to design experiments that concentrate on measuring those eight values. A related observation is that circularly polarized incident light [whose Stokes vector is invariant under **R** (ϕ)] produces scattered beams in all directions whose Stokes vector elements contain only the eight transformationally invariant matrix elements from the first and last columns of **M**.

5. CONCLUSIONS

In extending the notion of the Mueller matrix from its ordinary 2-D use to a 3-D application, we encountered a question of how to define reference frames. The Stokes vector describing a scattered beam of light is always referenced to its own scattering plane, the plane containing itself and the incident beam. However, two separate methods for referencing the incident beam itself are possible. In the first method leading to **D**, the incident Stokes vector "floats" in the sense that it is continuously redefined with respect to whichever scattering plane is under consideration at the moment. In the second method resulting in **M**, the incident beam's Stokes parameters are given fixed values that are tied to one particular selected reference plane. The first method is the most natural result of a calculation, lends itself to relatively simple graphical representations, and is most useful for characterizing a scattering particle. The second method is often the most natural result of an experiment, and the corresponding **M** matrix predicts the outgoing Stokes vectors, in all directions, resulting from given incident Stokes vectors. However, the **M** matrix is represented by graphical images whose complexity can mask the underlying symmetries of the scatterer. This report is intended to show that it is not a matter of deciding which reference system is "correct," but rather to choose the one more suitable for the task at hand.

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